# SUPPLEMENTARY INFORMATION (ESI)

# Underpinning the Interaction between NO<sub>2</sub> and CuO nanoplatelets at Room-Temperature By Tailoring Synthesis Reaction Base and Time

Dina N. Oosthuizen,\*, †, ‡ David E. Motaung,<sup>†, §</sup> André M. Strydom\*\* and Hendrik C. Swart†

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<sup>&</sup>lt;sup>+</sup> Department of Physics, University of the Free State, P. O. Box 339, Bloemfontein ZA9300, South Africa

<sup>&</sup>lt;sup>‡</sup> DST/CSIR National Centre for Nano-structured Materials, Council for Scientific Industrial Research, Pretoria, 0001, South Africa

<sup>&</sup>lt;sup>§</sup> Department of Physics, University of Limpopo, Private Bag X1106, Sovenga, 0727, South Africa

<sup>&</sup>lt;sup>\*\*</sup> Highly Correlated Matter Research Group, Department of Physics, University of Johannesburg, P.O. Box 524, Auckland Park, 2006, South Africa

#### 1. XRD data of the CuO products

XRD diffraction was used to study the phase purity of all as-prepared CuO products. The average crystallite size of the synthesized CuO nanoplatelets was calculated using a Debye-Scherrer formula <sup>1</sup>, and the lattice strain ( $\epsilon$ ) was calculated using the following relation:

$$\varepsilon = \frac{\beta_{2\theta} \cos \theta}{4} \tag{1}$$

where  $\theta$  is the diffraction angle and  $\beta_{2\theta}$  is the full width at half maximum intensity (FWHM) of the corresponding peak, as shown in Table S1.

**Table S1:** Summary of *d*-spacing, lattice strain and average crystallites size (D) estimated for **E-NaOH** to **E-NaOH-60**, **E-urea** to **E-urea-60** and **C-NH<sub>4</sub>OH** to **C-NH<sub>4</sub>OH-60**, with \* previously reported in Ref. 2.

	20 (°)	<i>d</i> -spacing (nm)	D (nm)	Lattice Strain (ε)			
NaOH as base							
* <b>E-NaOH 25 min</b> : 75 °C	35.65;	0.25159;	12.418;	0.0878;			
	38.85	0.23160	10.969	0.1609			
<b>E- NaOH-45:</b> 75 °C	35.63;	0.25177;	10.985;	0.0971;			
	38.81	0.23183	9.799	0.1824			
<b>E- NaOH-60:</b> 75 °C	35.59;	0.25201;	10.828;	0.0956;			
	38.76	0.23211	9.748	0.1860			
NH₄OH as base							
* <b>C-NH₄OH 25 min:</b> 55 °C	35.44;	0.2531;	16.147	0.0553;			
	38.57	0.2332	11.166	0.1707			
<b>C-NH₄OH-45</b> : 55 °C	35.49;	0.2527;	12.447	0.0755			
	38.57	0.2332	8.913	0.2136			
<b>C-NH₄OH-60:</b> 55 °C	35.48;	0.2527;	13.393	0.0699			
	38.59	0.2331	9.876	0.1921			
Urea as base							
* <b>E-Urea 25 min:</b> 75 °C	35.56;	0.2522;	10.18	0.0988;			
	38.74	0.2322	9.21	0.1977			
<b>E-Urea-45:</b> 75 °C	35.30;	0.2540;	10.758;	0.0704;			
	38.53	0.2334	9.049	0.2126			
<b>E-Urea-60:</b> 75 °C	35.37;	0.2535;	8.279;	0.1001;			
	38.58	0.2332	7.669	0.2480			

#### 2. Raman data of the CuO products

According to the Raman spectra, in Figure S1a, the intensity of the  $A_{1g}$ ,  $B_{1g}$  and  $B_{2g}$  peaks, for **E-NaOH-45** and **E-NaOH-60**, were the largest. A gradual increase in full width at half maximum (FWHM), indicating the increase in nanorods size, was observed for **E-NaOH-45** to **E-NaOH-60**. The Raman analyses of **C-NH<sub>4</sub>OH-45** and **C-NH<sub>4</sub>OH-60**, shown in Figure S1b, showed no significant peak shifts for the  $A_{1g}$  and  $B_{1g}$  modes; however a red shift was observed for the  $B_{2g}$  mode of **C-NH<sub>4</sub>OH-60**. The spectrum of **C-NH<sub>4</sub>OH-45** showed the highest observed peak intensity. The Raman spectrum of **E-Urea-45** showed a slight  $A_{1g}$  peak shift to larger wavenumbers, while the spectrum of **E-Urea-60** indicated a shift to larger wavenumbers of the  $B_{2g}$  mode, as displayed in Figure S1c. This red shift denotes that the grain size increases <sup>3</sup>, which is consistent with the XRD analyses.



**Figure S1:** The Raman spectra of the as-prepared CuO products (a) **E-NaOH-45** to **E-NaOH-90**, (b) **E-Urea-45** to **E-Urea-60** and (c) **C-NH<sub>4</sub>OH-45** to **C-NH<sub>4</sub>OH-60**.

# **3. BET surface analyses of the CuO products**

Table S2 shows the BET surface areas, pore volumes and pore sizes of the as- CuO products prepared at prolonged reaction times, **E-NaOH-45** to **E-NaOH-90**, **C-NH<sub>4</sub>OH-45** to **C-NH<sub>4</sub>OH-60**, **D-Urea-45** to **D-Urea-60** and **E-Urea-45** to **E-Urea-60**, as well as previously reported BET results.

**Table S2:** The results of the BET analysis of the as-prepared, **E-NaOH**, **C-NH**<sub>4</sub>**OH**, **D-Urea** and **E-Urea**, CuO products with prolonged reaction times. \*Data from Ref. 2.

Samples	BET surface area	Pore Volume	Pore Size					
	(m² g <sup>-1</sup> )	(cm <sup>3</sup> g <sup>-1</sup> )	(nm)					
NaOH as base								
*E-NaOH: 75 °C	55.76 ± 0.4	0.3471	29.59					
E-NaOH-45	64.85 ± 0.29	0.3701	19.33					
E-NaOH-60	67.61 ± 0.11	0.3898	20.59					
NH₄OH as base								
*C-NH₄OH: 55 °C	11.51 ± 0.10	0.0508	30.72					
C-NH₄OH-45	16.73 ± 0.15	0.0709	28.09					
C-NH₄OH-60	8.90 ± 0.15	0.0489	41.40					
Urea as base								
*E-Urea: 75 °C	55.62 ± 0.18	0.3101	19.10					
E-Urea-45	52.32 ± 0.10	0.2994	14.93					
E-Urea-60	56.05 ± 0.11	0.2900	17.75					



Figure S2: The BET surface area plots of E-NaOH-45 to E-NaOH-90, E-Urea-45 to E-Urea-60 and C-NH₄OH-45 to C-NH₄OH-60.

### 4. Hall-Effect analyses of the CuO products

Based on their gas sensing performance, some of the as-prepared CuO products were selected for Hall-Effect analyses. The as-prepared CuO products were dropcasted on a glass substrate to form CuO thin films which resulted to a thickness of ~ 0.5  $\mu$ m. The Hall Effect results showed that all the as-prepared CuO thin films were *p*-type, with the other electrical parameters summarized in Table S3.

 Table S3: Hall-Effect Analyses data of some of the as-prepared CuO products.

Samples	Carrier Type	Resistivity, ρ (x10³ Ω.cm)	Carrier concentration, n (x10 <sup>12</sup> cm <sup>-3</sup> )	Charge carrier mobility, μ (cm².V <sup>-1</sup> s <sup>-1</sup> )
E-NaOH-45	<i>p</i> -type	7.887	7.21	109.70
E-NaOH-60	<i>p</i> -type	2.884	9.82	220.40
E-Urea-60	<i>p</i> -type	3.471	4.94	363.90

Please note: Since the resistivity for powder films are high, the Hall-Effect results are only used to indicate the possible difference in carrier mobility and concentration etc. of the selected samples relative to observed gas sensing performance of the samples.

# 5. Gas sensing analyses of the CuO products

All the CuO sensing layers prepared from the synthetized CuO nanostructures were deposited with a drop coating technique. An overview and cross-section images of the **E-NaOH-45** sensor displays the average thickness of the sensing layers (see Figure S3a and b).



Figure S3: (a) Top view and (b) the cross-section image of the CuO sensing layer deposited from E-NaOH-45.



Figure S4: The sensor resistance corresponding calculated response, S of the E-NaOH-60 exposed to 40 ppm  $NO_2$  gas pulses over 12 hours at room temperature.

After the gas sensing system (also referred to as the gas testing station) was flushed with synthetic air at the room temperature, the duration of the gas cycles (i.e. pulse duration - gas on gas off cycles) was tested by exposing the sensors to a single test gas (in this instance 40 ppm NO<sub>2</sub> gas cycles). Figure S5 shows the exposure of sensor **E-NaOH-60** to a gas concentration (40 ppm) pulsed for 30 minutes, followed by synthetic air for 30 minutes, denoted as the first gas cycle, and subsequently followed by pulses with an interval of 20 and 10 minutes, respectively. Although, steady state was not reached during these cycles and drift in sensor resistance was observed, as seen in Figure S5, the large response to NO<sub>2</sub> gas at room temperature motivated the continuation of sensors' performance testing in humid conditions ranging from 5 to 60 % relative humidity.



**Figure S5:** The dynamic resistance curve of the **E-NaOH-60** exposed to 40 ppm  $NO_2$  gas pulses with varying gas cycle durations (i.e. gas on, gas off cycles – 30, 20 and 10 minutes).



Figure S6: The dynamic resistance curves of E-urea-60 in 40 ppm  $NO_2$  cycles in dry synthetic air and various RH (%) conditions at room temperature.

Figure S7a shows the real-time gas sensing response of **E-NaOH-60**-based gas sensor in CH<sub>4</sub> gas in dry air. As seen in Figure S7b, the CuO material prepared at 75°C in 60 minutes, showed the highest sensitivity to CH<sub>4</sub> gas, compared to the other material prepared at 75 °C. However, a large drift in sensor current was observed, which was attributed to the low rate of diffusion of gas molecules away from the surface of the sensor. As shown in Figure S7c, increased sensitivity to CH<sub>4</sub> gas was observed for **E-NaOH-45** and **E-NaOH-90**, with maximum sensitivities of around 41.5 ppm<sup>-1</sup>.



**Figure S7:** (a-b) The dynamic response curve of the **E-NaOH-60**, extracted from different  $CH_4$  concentration cycles, and (c) the  $CH_4$  response variation of **E-NaOH-45** and **E-NaOH-60**, to concentrations ranging from 10 to 40 ppm, at room temperature. \*Note: the lines visible in (c) are not experimental data; it is used to guide the reader.

Low response was showed when the **E-NaOH-60**-based gas sensor was subjected to four cycles of 40 ppm  $CH_4$  gas in more humid atmosphere (i.e. 20 to 60 % RH), as seen in Figure S8.



**Figure S8:** (a) Stacked dynamic sensor current curves of **E-NaOH-60** to four cycles of 40 ppm  $CH_4$  dosed in dry syn. air and various humid environments (i.e. 10 to 60% RH) at room temperature. (b) A plot of the calculated responses of **E-NaOH-60** to the 2<sup>nd</sup> cycle of  $CH_4$  gas with respect to the different RH %.

# References

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